

Pressure Study of the Noncentrosymmetric 5d-Electron Superconductors CaMSi_3 ($M = \text{Ir, Pt}$)

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We report a hydrostatic pressure study on the Rashba-type noncentrosymmetric superconductors CaMSi_3 ($M = \text{Ir, Pt}$). The temperature dependence of the resistivity of both compounds is well described by the conventional Bloch-Grüneisen formalism at each pressure. This fact suggests that electron-phonon scattering is dominant in these compounds. The superconducting critical temperature T_c decreases with pressure above 0.41 to 2 GPa at a rate of ~ 0.2 K/GPa for both compounds. This T_c behavior can be explained with a modest decrease in the density of states based on the conventional BCS theory.

Keywords: noncentrosymmetric superconductor, hydrostatic pressure, d -electron system, resistivity, superconducting H - T phase diagram

I. INTRODUCTION

Superconductors without the inversion symmetry have potentials to exhibit various novel phenomena and have recently been actively studied [1–17]. Studies of this new category of superconductors, which are called noncentrosymmetric superconductors (NCSCs), were initiated by the discovery of superconductivity in the heavy Fermion CePt_3Si [1]. The upper critical field H_{c2} of this compound is substantially larger than the Pauli limiting field, at which the ordinary spin-singlet superconductivity becomes unstable owing to the Zeeman energy. This large H_{c2} is attributable to a mixing between spin-singlet and spin-triplet states resulting from the fact that the parity is no longer a meaningful label for NCSCs. In addition to such parity violation, another important feature of NCSCs is the existence of the anisotropic spin-orbit interaction (ASOI), which may affect both the normal and superconducting states. Up to now, a number of NCSCs have been discovered and investigated, e.g., UIr [2], CeRhSi_3 [3], CeIrSi_3 [18], CeCoGe_3 [19], $\text{Li}_2(\text{Pd}_{1-x}\text{Pt}_x)_3\text{B}$ [4–7], Ru_7B_3 [8, 9], $\text{Mg}_{10}\text{Ir}_{19}\text{B}_{16}$ [10, 11], BaPtSi_3 [12], $\text{Mo}_3\text{Al}_2\text{C}$ [13, 14], and CaMSi_3 ($M = \text{Ir, Pt}$) [17]. Indeed, some of them exhibit unconventional behaviors: e.g., CeRhSi_3 and CeIrSi_3 have exceptionally high H_{c2} values, $\text{Li}_2\text{Pt}_3\text{B}$ exhibits a temperature-independent Knight shift through the superconducting transition, and $\text{Mo}_3\text{Al}_2\text{C}$ shows a non-BCS behavior in the specific heat and the nuclear-lattice relaxation rate $1/T_1$. However, a majority of the NCSCs behave conventionally. Theoretically, it is predicted that the existence of a strong electron correlation in addition to a strong ASOI is crucial for unconventional superconducting phenomena [15]. In fact, this prediction seems to explain the reported unconventional behavior in the cerium-based NCSCs, in which strong electron correlations originate from the interaction of conduction electrons with f -electrons. However, the cerium-based NCSCs reported up-to-date also exhibit antiferromagnetic ordering within or near the superconducting phase. This fact complicates the situation, because antiferromagnetic spin fluctuation without

parity violation can lead to non- s -wave Cooper pairing as in the cuprate superconductors. Thus, in order to extract the roles of parity violation and ASOI, studies of NCSCs without spin fluctuations are valuable.

CaMSi_3 ($M = \text{Ir, Pt}$) compounds have recently been reported to be nonmagnetic, fully gapped superconductors without a strong electron correlation [16, 17]. Thus, these compounds may serve as model materials for investigations of phenomena originating from their noncentrosymmetric crystal structure with a strong ASOI [20], because these compounds are not affected by any complications due to magnetism. The application of pressure is useful because effects of their electronic state variations can be investigated without introducing impurities by chemical substitution. For example, one may expect changes in superconducting properties due to the appearance/disappearance of an ASOI-split pair of Fermi surfaces. Here, we report the hydrostatic pressure dependence of the resistivity of CaMSi_3 ($M = \text{Ir, Pt}$) as well as its magnetic field dependence, and discuss the variation in superconducting behavior.

II. EXPERIMENTAL PROCEDURE

The arc-melted polycrystalline samples used in this study are from the same batch as the crystals used in ref. 17. The samples were shaped into blocks with the dimension of approximately $0.8 \times 0.8 \times 2$ mm³. The samples were placed in a Teflon capsule with a hydrostatic pressure medium (Daphne 7373) and mounted in a commercial piston-cylinder pressure cell (R&D Support Co., Ltd.). The pressure inside was determined from the Curie temperature of HoCo_2 [21], which was mounted inside the capsule together with the samples. Resistivity measurements with a conventional four-probe a.c. bridge technique under hydrostatic pressure were performed in a ³He cryostat (Cryogenic Ltd.) down to 0.35 K. The excitation current was 10 mA, and the frequency was 13.8 Hz.

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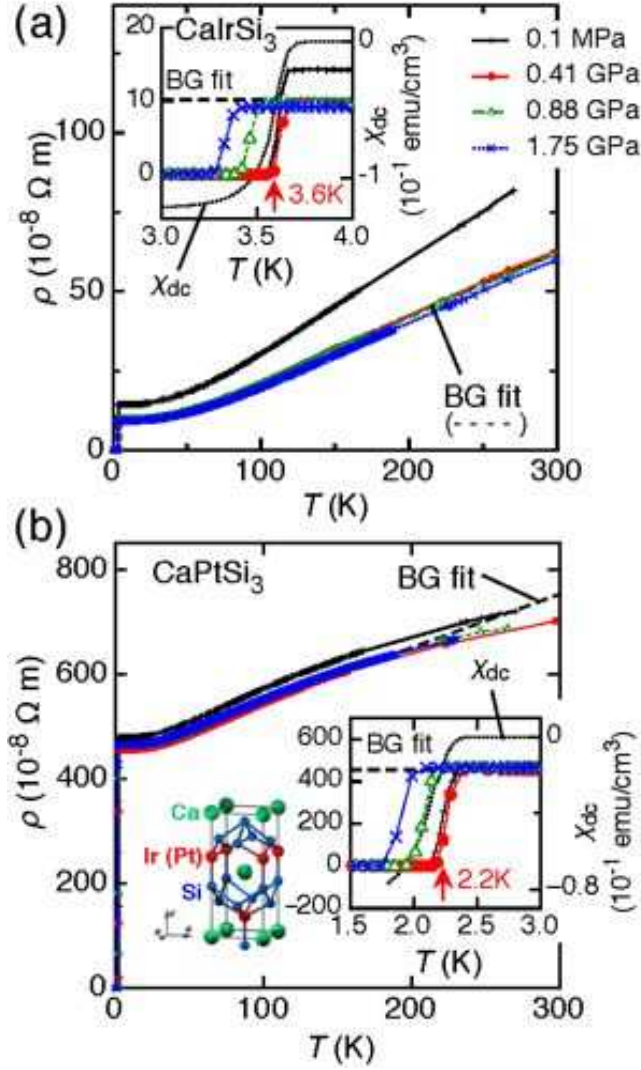


FIG. 1. (Color online) Temperature dependence of the resistivity of polycrystalline (a) CaIrSi_3 and (b) CaPtSi_3 for several pressures. Enlarged views near T_c are shown in each panel, and the zero-resistivity T_c values for 0 T and 0.41 GPa are indicated by arrows. The crystal structure is presented in the inset of (b). The fits of the resistivity for 0.41 GPa obtained by the Bloch-Grüneisen (BG) formula are also presented. The insets also present the dc susceptibility of the samples from which the crystals used in the present pressure study were cut. The susceptibility was measured under zero-field-cooled condition at 1 mT under ambient pressure. These data indicate that the samples for the present pressure study exhibit an almost 100% superconducting volume fraction.

III. RESULTS AND DISCUSSION

The temperature dependence of resistivity at several pressures is presented in Fig. 1, for which the sequence of the pressure application was in the order of 0.41 GPa, 0.88 GPa, 1.75 GPa, and 0.1 MPa. The superconducting transition temperature T_c is defined as the temperature at which the resistivity drops to 5% of its normal-state residual resistivity ρ_0 :

$\rho/\rho_0 = 5\%$. T_c values below 0.41 GPa are almost identical to the value in vacuum reported in ref. 17, and decrease with pressure for both compounds. Residual resistivity ratios $\rho_{300\text{K}}/\rho_0$ under pressure are ~ 4 for CaIrSi_3 and ~ 1.5 for CaPtSi_3 ; these values are also consistent with the previous report [17]. For both compounds, the resistivity at 0.1 MPa is larger than those at the other pressures. The difference does not seem to be intrinsic, i.e., it originates from the current path change by the pressure release. Each resistivity curve below 200 K is fitted by the conventional Bloch-Grüneisen (BG) formula

$$\rho = \rho_0 + \left(\frac{C}{\theta_D} \right) \left(\frac{T^5}{\theta_D^5} \right) \int_0^{\theta_D/T} \frac{x^5}{(\exp[x] - 1)(1 - \exp[-x])} dx$$

with three fitting parameters: C is a temperature-independent material constant describing the electron-phonon interaction, θ_D is the Debye temperature, and ρ_0 is the residual resistivity. The fitting for the 0.41 GPa data yields $\theta_D = 322$ K for CaIrSi_3 , which is consistent with $\theta_D = 360$ K deduced from the specific heat below 5 K [17]. This result suggests that the temperature dependence of resistivity is dominated by the electron-phonon scattering. However, we obtain $\theta_D = 171$ K for CaPtSi_3 , which is substantially smaller than the value from the specific heat (370 K) [17]. This difference in θ_D indicates that phonons having lower frequencies mainly contribute to the electron-phonon scattering. In addition, the resistivity tends to saturate more strongly than the BG approximation in the high-temperature region, as often observed in intermetallic compounds (e.g., ref. 22).

The magnetic field dependence of resistivity at several pressures is presented in Fig. 2. The direction of the magnetic field was parallel to the applied current. At each magnetic field, the overall temperature dependence for different pressures was similar to each other. This indicates that the electronic states as well as the scattering processes do not change much with pressure. As presented in Fig. 2, a clear field-dependent two-step transition was observed only for CaIrSi_3 which, additionally, exhibited a substantial broadening of the superconducting transition. This broadening indicates a spatial distribution of the superconducting fraction. Slight upturns just above the resistivity drop were observed for CaPtSi_3 in addition to the broadening of the transition. Such upturns have been observed and attributed to a pressure-induced charge-density-wave (CDW) ordering in Mo_3Sb_7 [23]. However, in the case of CaPtSi_3 , the upturn seems to be related to the superconducting transition because the onset temperature of this anomaly is suppressed by the field. Furthermore, the onset temperature is almost identical to the superconducting onset T_c reported in ref. 17. For these reasons, the upturns are unlikely associated with a CDW; rather, they are caused by the current path change or nontrivial vortex dynamics related to superconductivity.

The H - T phase diagrams deduced from the resistivity are presented in Fig. 3. Here, we show T_c defined as the 5% criterion (large symbols) and T_c^{onset} defined as the temperature at which the resistivity drops to 95% of ρ_0 (small symbols). For CaPtSi_3 under magnetic fields, T_c^{onset} is defined at the resistivity maximum in the transition region. The large deviation in

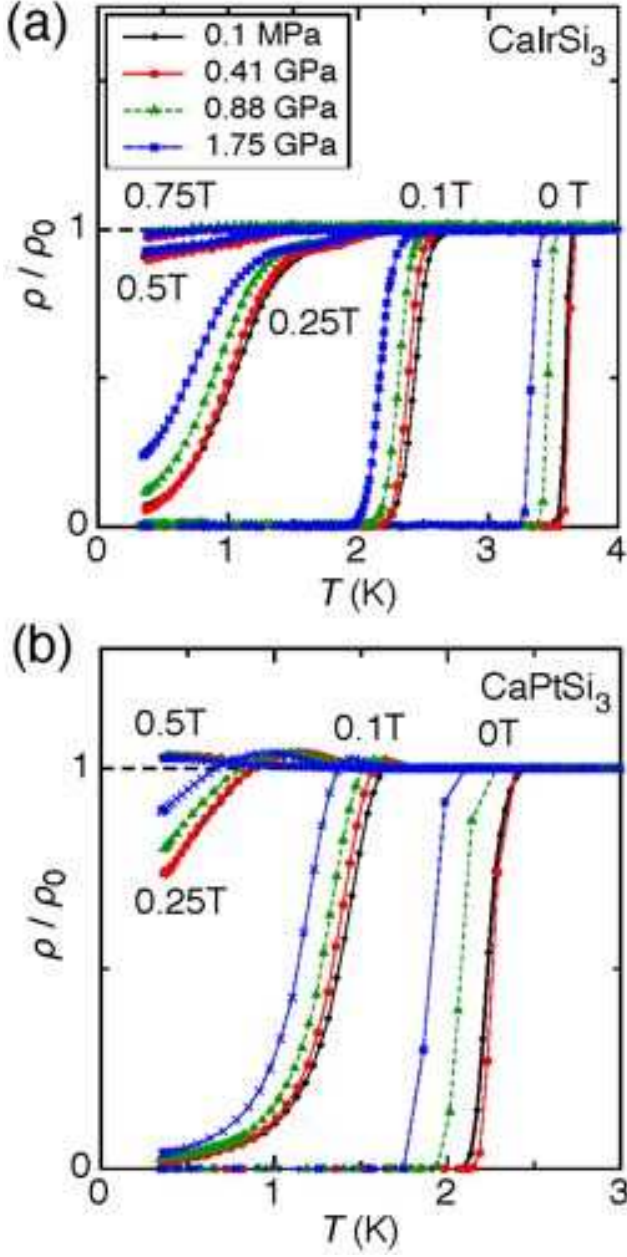


FIG. 2. (Color online) Magnetic field dependence of the low-temperature resistivity under pressure: (a) CaIrSi_3 and (b) CaPtSi_3 . Each data is normalized by its residual resistivity ρ_0 obtained by Bloch-Grüneisen fitting.

T_c from T_c^{onset} reflects the broadening of the transition. T_c^{onset} under zero pressure is taken from the previous report [17]. The H - T curves of the onset T_c under pressure have good overall similarity to those reported in ref. 17.

The pressure variations of T_c , θ_D determined by the Bloch-Grüneisen formula, and $H_{c2}(0)$ deduced from the linear extrapolation of T_c are presented in Fig. 4. A clear decrease in T_c is observed for both compounds above 0.41 GPa with a rate of approximately $dT_c/dP \sim -0.2$ K/GPa. The variation in T_c below 0.41 GPa is within the experimental precision. In

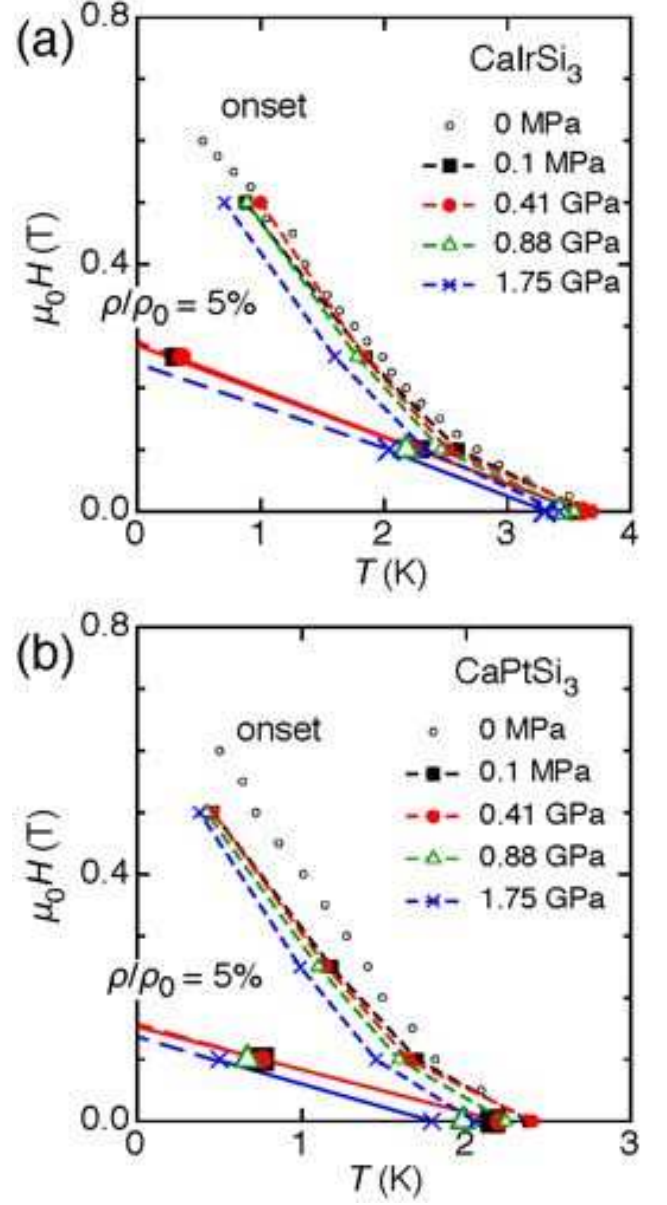


FIG. 3. (Color online) H - T phase diagram under pressure: (a) CaIrSi_3 and (b) CaPtSi_3 . The H - T curves at 0 MPa are taken from the previous report [17]. The large and small symbols indicate T_c and T_c^{onset} , respectively. See text for the definitions of T_c and T_c^{onset} .

contrast, θ_D does not exhibit any systematic change.

As discussed above, the electron-phonon and impurity scattering processes are dominant in both CaIrSi_3 and CaPtSi_3 . No other interactions that could give rise to Cooper pairing, such as spin fluctuations, have been reported for these compounds. Therefore, pairing by the conventional phonon-mediated attractive interaction is most probably realized. Furthermore, specific heat is consistent with the weak-coupling BCS behavior [17]. In the conventional weak-coupling BCS theory, T_c is given by $T_c = 1.13\theta_D \exp[-1/(N(0)V)]$, where $N(0)$ is the density of states at the Fermi level, and V is the magnitude of the interelectron attractive interaction. Note that

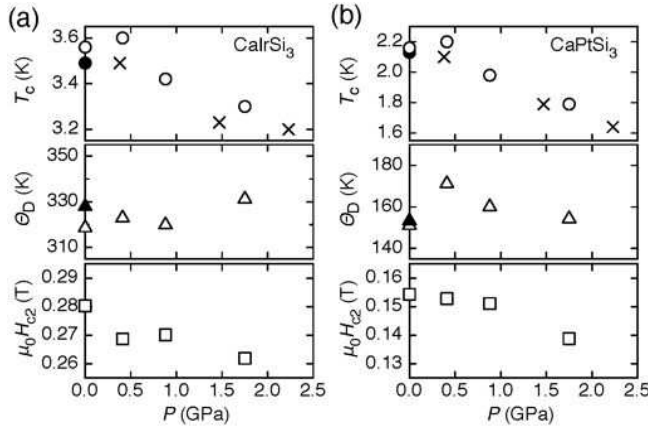


FIG. 4. Pressure dependence of superconducting transition temperature T_c , Debye temperature Θ_D , and the upper critical field $H_{c2}(0)$ of (a) CaIrSi_3 and (b) CaPtSi_3 , presented by open symbols. The values represented by the closed symbols are deduced from the data reported in ref. 17, and the T_c values represented by crosses are taken from a test measurement using the same samples.

the formula indicates that T_c is an increasing function of Θ_D , $N(0)V$. As mentioned above, the variation in Θ_D does not reflect the variation in T_c with pressure, and therefore should not dictate the observed pressure dependence of T_c . As a result, the change in T_c with pressure likely originates from the change of $N(0)V$. In particular, the decrease in T_c with pressure above 0.41 GPa is probably due to the broadening of the bandwidth leading to the decrease in $N(0)$.

$H_{c2}(0)$ presented in Fig. 4 tends to decrease with pressure. This tendency is clearly observed from the raw data in Fig. 2. The Ginzburg-Landau (GL) coherence length $\xi(0)$ values, calculated from the relation $\mu_0 H_{c2}(0) = \Phi_0 / 2\pi \xi^2(0)$, where Φ_0 is the flux quantum, are approximately 35 nm for CaIrSi_3 and 46 nm for CaPtSi_3 at 0.41 GPa with no significant change in the presented pressure range. This estimated $\xi(0)$ is an effective value given by $\xi = (1/\xi_0 + 1/l)^{-1}$, where ξ_0 is the intrinsic GL coherence length, and l is the mean free path. Therefore, the change in effective coherence length can be explained by a change in either ξ_0 or l . Considering the fact that l is determined by the mean interimpurity distance at low temperatures, l should be independent of pressure. The pressure dependence of $\mu_0 H_{c2}$ is thus explained by the variation in ξ_0 , which is reflected in the variation in ξ .

Our study indicates that T_c decreases by about 20% with pressure (1.75 GPa) without changes in the behavior of the

resistivity nor the shape of the $H_{c2}(T)$ curve for both compounds. This indicates that the electronic states do not exhibit any substantial change in the examined pressure range, except for a modest decrease in $N(0)V$. However, the band calculations have revealed that several edges of ASOI-split bands are located in the vicinity of the Fermi energy [20]. Thus, it is expected that one can induce a substantial change in the electronic structure if one can shift the band edges across the Fermi energy by applying higher pressure. Furthermore, when one Fermi surface of the ASOI-split pair disappears, a topological superconducting state might be realized. Indeed, the creation of a topological superconducting state by the disappearance of one part of ASOI-split Fermi surfaces has been proposed in a slightly different context [24]. A study of the uniaxial pressure effect using a single crystal would also be favorable for investigating the relationship between the electronic state and the Rashba-type ASOI.

IV. CONCLUSIONS

We have investigated the hydrostatic pressure dependence of the resistivity and the superconducting behavior of the noncentrosymmetric superconductors CaIrSi_3 and CaPtSi_3 . The temperature dependence of resistivity is explained by the conventional Bloch-Grüneisen formalism, suggesting that the electron-phonon scattering is predominant in these compounds. The decrease in T_c for $P > 0.41$ GPa up to the maximum pressure in the present study can be explained by the decrease in $N(0)V$ within the conventional BCS theory. The magnetic field dependence does not exhibit any qualitative change with pressure.

V. ACKNOWLEDGMENT

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